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ance Notes on Codes and Abbreviations" appearing at the begin-
ning of each regular issue of the PCT Gazette.

(54) Title: FLAME-RETARDANT POLYPROPYLENE RESIN COMPOSITION

(57) Abstract: The present invention relates to flame-retardant polypropylene resin composition having improved mechanical prop-
erty and flame-retardant property. More specifically, the polypropylene resin composition comprises polypropylene resin, brome-
based flame-retardant agent, inorganic flame-retardant agent, antimony compound and chlorinated polyethylene.



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FLAME-RETARDANT POLYPROPYLENE RESIN COMPOSITION**Technical Field**

The present invention relates to novel flame-retardant
5 polypropylene resin compositions, which exhibit high
incombustibility while being fine in other mechanical
properties such as impact resistance, etc. More
specifically, the present invention relates to
incombustible polypropylene resin compositions comprising
10 an organic flame retardant, an inorganic flame retardant
and an antimony compound besides the major component,
polypropylene resin in specific ratios.

Background Art

15 Since they have good processability, chemical
resistance, weather resistance and hinge properties,
polypropylene resins have been widely used, in the form of
injection-molding products, films or blow-molding products,
in various fields such as parts of automobiles, building
20 materials, parts of electrical appliances, etc. However,
they are essentially inflammable, so they need to be
supplemented with a variety of organic or inorganic flame
retardants in order to obtain incombustibility. Generally,
an organic or inorganic flame retardant in combination with
25 antimony trioxide is used to make polypropylene resins
incombustible.

Exemplary incombustible polypropylene resin
compositions containing such flame retardants include
compositions prepared by adding to a polypropylene resin a
30 bromine-based flame retardant such as
decabromodiphenylether, bis(pentabromophenyl)ethane,

etc.(See: Japanese Laid-open Publication Nos. HEI 8-120126 and HEI 7-76640); compositions prepared by adding to a polypropylene resin a tetrabromo bisphenol A-based or tetrabromo bisphenol S-based flame retardant(See: Japanese Laid-open Publication No. HEI 8-302102); compositions prepared by adding to a polypropylene resin a hydrated inorganic compound such as magnesium hydroxide, aluminum hydroxide or hydrotalcite(See: Japanese Laid-open Publication Nos. 53-92855, 54-29350, 54-77658, 56-25954, 57-87462 and 60-110738); compositions prepared by adding to a polypropylene resin a polyethylene resin with a melt index of 0.01~2.0, a halogen compound(e.g., decabromodiphenyl ether or dodecachloro-dodecahydromethanodibenzocyclooctene), and an inorganic filler selected from the group consisting of talc powder, kaolin, celestite, silica and diatomite(See: Japanese Laid-open Publication No. 55-30739); and compositions prepared by adding to a polypropylene resin either a reaction product, which is generated from a reaction between an aldehyde compound and a nitrogenous compound, wherein $=C=O$ (or $=C=S$ or $=NH$) group is introduced into a ring structure of ammonium phosphate(or amine phosphate), or an oligomer(or polymer) of 1,3,5-triazine derivatives(See: Japanese Laid-open Publication Nos. 52-146452 and 59-147050).

However, the compositions, which were prepared by adding an inorganic compound such as magnesium hydroxide to a polypropylene resin in order to obtain a highly incombustible resin composition, were found to be poor in plasticity. On the other hand, the compositions prepared by adding a decabromophenyl-based compound to a

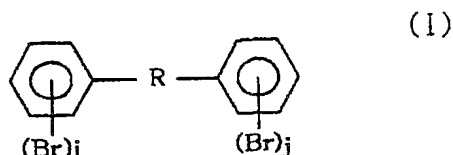
polypropylene resin are not very poor in plasticity, but their impact resistance is very low. Furthermore, though they exhibit the incombustibility of V-0 grade at 1/32-inch thickness when tested according to the vertical firing test protocol out of "COMBUSTIBILITY TESTS OF PLASTIC MATERIALS FOR MACHINE PARTS" of UL Subject 94 (Underwriters Laboratories Incorporation), these compositions are unsuccessful to show higher incombustibility corresponding to 5V grade. Therefore, they are unsuitable to be used in any parts requiring the high incombustibility of 5V grade as well as excellent impact and thermal resistance.

Disclosure of the Invention

A feature of the present invention is to solve these problems of the prior arts and to provide novel incombustible polypropylene resin compositions showing the high incombustibility of 5V grade in a state of thin plate, and, at the same time, showing excellent impact resistance and thermal resistance, by adding, to a polypropylene resin, i) a bromine-based flame retardant having a specific structure, ii) an antimony compound as a synergist of the flame retardant, iii) a magnesium hydroxide as an inorganic flame retardant, and iv) a chlorinated polyethylene as both a modifier of impact resistance and an auxiliary flame retardant.

According to the present invention, there is provided incombustible polypropylene resin compositions comprising (A) 31~61 wt% of a polypropylene resin, (B) 13~35 wt% of a bromine-based flame retardant represented by the following formula (I), (C) 3~21 wt% of an antimony trioxide, (D) 3~30 wt% of a magnesium hydroxide, and (E) 2~15 wt% of a

chlorinated polyethylene:



(in the above formula (I), R is oxygen atom or $\text{C}_{1\sim 6}$ alkyl group; i is an integer of 0~5; and j is an integer of 1~5)

As used herein, "polypropylene" refers to propylene homopolymer as well as propylene-olefin block copolymer containing 50 wt% or more polypropylene unit.

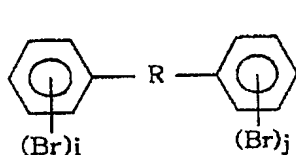
In the incombustible resin compositions of the present invention, the polypropylene resin (A) may be a crystalline polypropylene homopolymer, or a crystalline copolymer consisting of propylene and one or more compounds selected from the group consisting of ethylene, 1-butene, 1-pentene, 1-hexene, 4-methylpentene, 1-heptene, 1-octene and 1-decene. Melt index of the polypropylene resin is in the range of 0.1~60g/10min, preferably 0.3~40g/10min. If the melt index of the polypropylene resin is below 0.1g/10min, excessive pressure is loaded during injection molding and flow marks occur on the surface of the molding products, and thus defect in appearance is resulted. Conversely, if the melt index of the polypropylene resin is over 60g/10min, impact resistance of the molding products becomes lower suddenly and drip of ignitable melted material is likely to occur.

According to the present invention, any of the conventional crystalline polypropylene resins can be used regardless of their stereoregularity. Nevertheless, a crystalline polypropylene showing an isotactic pentad

fraction of preferably 0.80~0.99, more preferably 0.85~0.99, and most preferably 0.90~0.99 in ^{13}C -NMR(nuclear magnetic resonance) spectrum is preferred.

In the resin compositions of the present invention, amount of the polypropylene resin (A) is 31~61% by weight, and preferably 35~58% by weight. If the polypropylene resin content is below 31% by weight, plasticity and impact resistance of the resin composition decrease in spite of high thermal resistance. Conversely, if the polypropylene resin content is over 61% by weight, thermal resistance and incombustibility of the resin composition decrease in spite of good plasticity and impact resistance, and therefore the composition cannot be used in parts requiring all of thermal resistance, incombustibility and impact resistance.

In the present invention, the bromine-based flame retardant (B) can be represented by the following formula (I), and preferably contains 30 wt% or more bromine:



(in the above formula (I), R is oxygen atom or $\text{C}_1\sim\text{C}_6$ alkyl group; i is an integer of 0~5; and j is an integer of 1~5)

In the above formula (I), the alkyl group is preferably methyl or ethyl group.

In the resin compositions of the present invention, amount of the bromine-based flame retardant (B) is 13~35% by weight. If the bromine-based flame retardant content is below 13% by weight, flameproof effect is insufficient to obtain the incombustibility corresponding to UL94 5V

grade. Conversely, if the bromine-based flame retardant content is over 35% by weight, overall mechanical properties of the composition, particularly thermal resistance and tensile strength are spoiled, and electrical
5 properties are also lowered.

In the present invention, the antimony trioxide (C) used as an auxiliary flame retardant is commercially available. Any antimony trioxide in powder form can be used, wherein mean particle size of the powder, preferably,
10 ranges from 0.2 to 5.0 μ m.

In the resin compositions of the present invention, amount of the antimony trioxide (C) is 3~21% by weight. If the antimony trioxide is contained below 3% by weight, synergistic effect with the bromine-based flame retardant
15 is too low to achieve the incombustibility of UL94 5V grade. On the contrary, if the antimony trioxide is contained over 21% by weight, there is no more increase in the synergistic effect and impact resistance is significantly declined.

The antimony trioxide (C) and the bromine-based flame
20 retardant (B) show a synergism in improving incombustibility. Especially, when the antimony trioxide is 1/6~2 times as much as the bromine-based flame retardant, this synergism is optimized.

The magnesium hydroxide (D) used as an inorganic flame
25 retardant according to the present invention plays an important role in the composition. That is, the magnesium hydroxide absorbs the heat of reaction in the event of combustion of the resin to be decomposed into moisture. This, together with the synergism between the bromine-based
30 flame retardant and the antimony trioxide as described above, enables the incombustibility of the resin

composition to be highly improved. Moreover, the magnesium hydroxide prevents ignitable melted material from dripping during combustion, so that generation of dripping substances, such as those defined in the incombustibility grade 5V according to "UL94 COMBUSTIBILITY TEST", can be avoided.

In the resin compositions of the present invention, amount of the magnesium hydroxide (D) is 3~30% by weight. If the magnesium hydroxide content is below 3% by weight, the heat of combustion is not sufficiently absorbed, so that the synergistic effect on improvement of incombustibility is little, and further the drip of the melted material is not properly prevented. Conversely, if the magnesium hydroxide content is over 30% by weight, the composition is poor in fluidity and the other mechanical properties such as impact resistance and tensile strength, and its plasticity is also lowered.

As the inorganic flame retardant used in the present invention, an agent whose surface is treated with a silane-based coupling agent or with a metal stearate is preferred. In the case that untreated one is used, the resin composition is poor in mechanical properties including impact resistance.

The chlorinated polyethylene (E) used in the present invention is a polyethylene wherein some of hydrogen atoms are substituted with chlorine atoms. Thus chlorinated polyethylene has a characteristic of rubber rather than that of the usual polyethylene itself, and so it acts as both a modifier of impact resistance and a flame retardant. The chlorinated ethylene improves impact resistance of the resin composition, and also improves incombustibility of

the resin composition through providing the halogen element to the composition.

In the resin compositions of the present invention, the chlorinated polyethylene used as an impact-resistant stiffener and an auxiliary flame retardant contains preferably 20~50 wt% of chlorine, and its amount is 2~15% by weight. If chlorine content of the chlorinated polyethylene is below 20 wt%, effect on improvement of impact resistance is slight. On the contrary, chlorine content of the chlorinated polyethylene is over 50 wt%, thermal degradation readily occurs during processing because of inherent low thermal stability of the chlorinated polyethylene. Meanwhile, if the chlorinated polyethylene content is below 2 wt% in the composition, sufficient reinforcing effect against impact can not be expected. Conversely, if the chlorinated polyethylene content is over 15 wt%, thermal stability significantly decreases.

To the incombustible resin compositions of the present invention may be added supplementary additives in order to further improve the mechanical properties such as stiffness and so on and thermal resistance of the composition. For example, one or more inorganic fillers selected from the group consisting of talc, silica, clay, kaolin, glass fiber, calcium carbonate and mica can be used solely or in combination.

According to the present invention, molding products made of the polypropylene resin can be made by mixing and mulling all of the components recited above to provide an incombustible polypropylene resin composition, and then molding the composition by any molding method such as

injection molding, extrusion molding, etc.

The polypropylene resinous molding products obtained from the above possess excellent impact resistance and thermal resistance as well as high incombustibility
5 corresponding to UL94 5V grade, and thus they are useful as parts of electrical or electronic appliances, parts of automobiles, and building materials. This is due to that: firstly, the bromine-based flame retardant and the antimony trioxide, in combination, provide a synergistic flame-
10 retardant effect, secondly, the inorganic flame retardant, i.e., magnesium hydroxide grants thermal resistance and further acts as another synergistic flame retardant, and finally, the chlorinated polyethylene provides impact-resistant and flame-retardant effects.

15 The incombustible resin compositions of the present invention can be prepared by the process as follows: polypropylene resin (A), bromine-based flame retardant (B) represented by the above formula (I), antimony trioxide (C), magnesium hydroxide (D) as an inorganic flame retardant,
20 chlorinated polyethylene (E), and the above mentioned various additives are filled in an agitating and mixing apparatus(e.g., Hensel mixer(brand name), super mixer or tumbler mixer) in specified amounts, and mixed with agitating for 1~10min, and then melted and mulled at
25 180~270°C by the use of a rolling mill or an extruder to provide pellets.

Best Mode for Carrying Out the Invention

The present invention can be more clearly understood
30 with referring to the following examples. It should be understood that the following examples are not intended to

restrict the scope of the present invention in any manner.

Protocols for Measuring Mechanical Properties

1) Izod impact strength

5 Izod impact strength was measured according to the test standard ASTM D256 by using test pieces(3.2mm in thickness) produced by injection molding.

2) Heat deflection temperature

Heat deflection temperature was measured according to
10 the test standard ASTM D648 under 4.6kgf load by using test pieces(127.0mm in length, 12.7mm in width, 6.4mm in thickness) produced by injection molding.

3) Incombustibility

Incombustibility was measured according to the
15 vertical(5V) and horizontal flat plane test protocols out of "COMBUSTIBILITY TESTS OF PLASTIC MATERIALS FOR MACHINE PARTS" of UL Subject 94. Test pieces of 1/32-inch thickness were used.

20 Example 1

In a Hensel mixer(brand name) were filled 4.4kg of crystalline ethylene-propylene block copolymer having a melt index(flow amount of melted resin measured for 10min at 230°C under a load of 2.16kg) of 3g/10min as a
25 polypropylene resin, 2.1kg of bis(pentabromophenyl)ethane[brand name: S-8010; Albermale Co., Ltd., USA] as a bromine-based flame retardant, 700g of antimony trioxide[brand name: SW; Ilsung Antimon Co., Ltd.], 2.0kg of magnesium hydroxide[brand name: Kisuma5A;
30 Kyowa Chemicals Co., Ltd., Japan] as an inorganic flame retardant, 800g of chlorinated polyethylene[brand name:

Tyryn 3611P; Dow Chemicals, Co., Ltd.], and 10g of calcium stearate as an additive, and then mixed for 3min with agitating. The resulting mixture was melted and extruded through an extruder of 30mm-caliber at 180~260°C to provide
5 pellets.

Examples 2~4 and Comparative examples 1~4

The same components as in Example 1 were filled in a Hensel mixer (brand name), except varying the respective
10 amount of the polypropylene resin, the bromine-based flame retardant and the magnesium hydroxide as shown in Tables 1 and 2. Then, they were mixed with agitating, and the resulting mixture was melted and extruded under the same condition as in Example 1 to provide pellets.

15 The pellets obtained from Examples 1~4 and Comparative examples 1~4 were dried for 3hrs at 100°C, and then molded through an injection molding machine, wherein maximum temperature of its cylinder had been adjusted to 230°C, to provide test pieces. The test pieces were subjected to
20 the tests for incombustibility and other mechanical properties as described above. The results are shown in Tables 1 and 2.

As can be seen from Tables 1 and 2, all of Examples 1~4, which were prepared according to the present invention,
25 showed high Izod impact strength and heat deflection temperature, and also showed high incombustibility corresponding to UL94 5V grade. On the contrary, all of Comparative examples 1~4 failed to concurrently show high impact resistance, thermal resistance and incombustibility
30 of 5V grade.

Example 5

In a Hensel mixer (brand name) were filled 4.1kg of crystalline ethylene-propylene block copolymer having a melt index (flow amount of melted resin measured for 10min at 230°C under a load of 2.16kg) of 3g/10min as a polypropylene resin, 2.1kg of bis(pentabromophenyl)ethane [brand name: S-8010; Albermale Co., Ltd., USA] as a bromine-based flame retardant, 1.0kg of antimony trioxide [brand name: SW; Ilsung Antimon Co., Ltd.], 2.0kg of magnesium hydroxide [brand name: Kisuma5A; Kyowa Chemicals Co., Ltd., Japan] as an inorganic flame retardant, 800g of chlorinated polyethylene [brand name: Tyrin 3611P; Dow Chemicals, Co., Ltd.], and 10g of calcium stearate as an additive, and then mixed for 3min with agitating. The resulting mixture was melted and extruded through an extruder of 30mm-caliber at 180~260°C to provide pellets.

Examples 6~8 and Comparative examples 5~7

The same components as in Example 5 were filled in a Hensel mixer (brand name), except varying the respective amount of the polypropylene resin, the bromine-based flame retardant, the antimony trioxide, the magnesium hydroxide and the chlorinated polyethylene as shown in Tables 1 and 2. Then, they were mixed with agitating, and the resulting mixture was melted and extruded under the same condition as in Example 5 to provide pellets.

The pellets obtained from Examples 6~8 and Comparative examples 5~7 were dried for 3hrs at 100°C, and then molded through an injection molding machine, wherein maximum temperature of its cylinder had been adjusted to 230°C, to

provide test pieces. The test pieces were subjected to the tests for incombustibility and other mechanical properties as described above. The results are shown in Tables 1 and 2.

5 As can be seen from Tables 1 and 2, all of Examples 6~8, which were prepared according to the present invention, showed high Izod impact strength and heat deflection temperature, and also showed high incombustibility corresponding to UL94 5V grade. In contrast, Comparative
10 example 5, wherein the chlorinated polyethylene content is below 2% by weight, failed to achieve the incombustibility of 5V grade, because effect of the chlorinated polyethylene on improvement of incombustibility was insufficient. On the other hand, Comparative example 6, wherein the
15 chlorinated polyethylene content is over 15% by weight, succeeded in achieving the incombustibility of 5V grade, but it was defective in that thermal resistance was abruptly decreased.

20 Example 9

In a Hensel mixer (brand name) were filled 4.4kg of crystalline ethylene-propylene block copolymer having a melt index (flow amount of melted resin measured for 10min at 230°C under a load of 2.16kg) of 3g/10min as a
25 polypropylene resin, 2.1kg of decabromo diphenyl ether [brand name: S-102E; Albermale Co., Ltd., USA] as a bromine-based flame retardant, 700g of antimony trioxide [brand name: SW; Ilsung Antimon Co., Ltd.], 2.0kg of magnesium hydroxide [brand name: Kisuma5A; Kyowa
30 Chemicals Co., Ltd., Japan] as an inorganic flame retardant, 800g of chlorinated polyethylene [brand name: Tyrin 3611P;

Dow Chemicals, Co., Ltd.], and 10g of calcium stearate as an additive, and then mixed for 3min with agitating. The resulting mixture was melted and extruded through an extruder of 30mm-caliber at 180~260°C to provide pellets.

5

Comparative examples 8~9

The same components as in Example 9 were filled in a Hensel mixer (brand name), except varying the amount of the polypropylene resin and the kind of the bromine-based flame retardant as shown in Table 2. Then, they were mixed with agitating, and the resulting mixture was melted and extruded under the same condition as in Example 9 to provide pellets.

The pellets obtained from Comparative examples 8~9 were dried for 3hrs at 100°C, and then molded through an injection molding machine, wherein maximum temperature of its cylinder had been adjusted to 230°C, to provide test pieces. The test pieces were subjected to the tests for incombustibility and other mechanical properties as described above. The results are shown in Table 2.

As can be seen from Tables 1 and 2, Example 9, which was prepared according to the present invention, showed high Izod impact strength and heat deflection temperature, and also showed high incombustibility corresponding to UL94 5V grade. However, in case that the bromine-based flame retardant represented by the above formula (I) was replaced with tetrabromo bisphenol A-based or tetrabromo bisphenol S-based flame retardant, the incombustibility of 5V grade was not achievable.

30

Table 1

(unit: 100g)

	Ex. 1	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	Ex. 8	Ex. 9
(A)	44	61	40	57	41	51	31	37	44
(B)-1	21	21	35	13	21	18	21	21	-
(B)-2	-	-	-	-	-	-	-	-	21
(C)	7	7	7	7	10	3	21	7	7
(D)-1	20	10	10	8	20	20	25	20	20
(E)	8	8	8	15	8	8	2	15	8
Izod impact strength (kg·cm/cm)	8.0	9.2	6.2	9.1	10.4	6.4	8.3	13.4	9.2
Heat deflection Temperature (°C)	132	126	129	125	124	133	130	121	125
Incombustibility (UL94)	5V	5V	5V	5V	5V	5V	5V	5V	5V

*Ex.: Example

Table 2

(unit: 100g)

	Com.	Com.	Com.	Com.	Com.	Com.	Com.	Com.	Com.
	1	2	3	4	5	6	7	8	9
(A)	62.5	29	38	26	51.5	32	44	44	44
(B)-1	21	21	11	37	21	21	21	-	-
(B)-3	-	-	-	-	-	-	-	21	-
(B)-4	-	-	-	-	-	-	-	-	21
(C)	7	7	11	7	7	7	7	7	7
(D)-1	1.5	35	20	20	20	20	-	20	20
(D)-2	-	-	-	-	-	-	20	-	-
Izod impact strength (kg·cm/cm)	9.6	4.0	11.9	4.0	4.5	12.8	4.9	13.5	12.9
Heat deflection Temperature (°C)	125	135	120	128	129	100	138	110	115
Incombustibility (UL94)	completely burned	5V	completely burned	5V	completely burned	5V	completely burned	drip	drip

*Com.: Comparative example

5 [Notes]

•Component (A): polypropylene resin[brand name: BJ300; Samsung General Chemicals Co., Ltd.]

•Component (B)-1: bromine-based flame retardant, bis(pentabromophenyl)ethane[brand name: S-8010; Albermale Co., Ltd., USA]

•Component (B)-2: bromine-based flame retardant, decabromo diphenyl ether[brand name: S-102E; Albermale Co., Ltd., USA]

•Component (B)-3: bromine-based flame retardant, tetrabromo bisphenol A-bis(2,3-dibromopropyl ether)[brand name: PE68; Great Lakes Co., Ltd., USA]

•Component (B)-4: bromine-based flame retardant, tetrabromo

bisphenol S-based[brand name: Nonnen52; Marubishi Chemicals Co., Ltd., Japan]

•Component (C): antimony trioxide[brand name: SW; Ilsung Antimon Co., Ltd.]

5 •Component (D)-1: magnesium hydroxide[brand name: Kisuma5A; Kyowa Chemicals Co., Ltd., Japan]

•Component (D)-2: Talc[brand name: KCN5200; Koshi Co., Ltd., Japan]

10 •Component (E): chlorinated polyethylene[brand name: Tyrin 3611P; Dow Chemicals Co., Ltd.]

Industrial Applicability

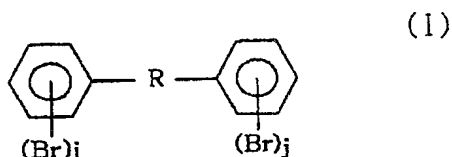
As can be seen in the above Examples and Comparative examples, the polypropylene resin compositions of the present invention have excellent impact resistance and thermal resistance, and also exhibit high incombustibility corresponding to UL94 5V grade at even the thickness of 1/32 inch. Therefore, they are useful for manufacture of parts of electrical appliances and automobiles, building materials, interior decorative materials, etc., in which these properties are required.

15

20

Claims

1. An incombustible polypropylene resin composition comprising (A) 31~61 wt% of a polypropylene resin, (B) 13~35wt% of a bromine-based flame retardant represented by the following formula (I), (C) 3~21 wt% of an antimony trioxide, (D) 3~30 wt% of a magnesium hydroxide, and (E) 2~15 wt% of a chlorinated polyethylene:



10

(in the above formula (I), R is oxygen atom or $\text{C}_{1\sim6}$ alkyl group; i is an integer of 0~5; and j is an integer of 1~5).

2. The incombustible polypropylene resin composition according to claim 1, wherein the polypropylene resin is a crystalline polypropylene homopolymer, or a crystalline copolymer consisting of propylene and one or more compounds selected from the group consisting of ethylene, 1-pentene, 1-hexene, 4-methylpentene, 1-heptene, 1-octene and 1-decene.
3. The incombustible polypropylene resin composition according to claims 1 or 2, wherein melt index of the polypropylene resin is in the range of 0.1~60g/10min.
4. The incombustible polypropylene resin composition according to claims 1 or 2, wherein isotactic pentad fraction of the polypropylene resin is in the range of 0.80~0.99.
5. The incombustible polypropylene resin composition

according to claim 1, wherein bromine content of the bromine-based flame retardant is 30 wt% or more.

6. The incombustible polypropylene resin composition according to claim 1, wherein chlorine content of the chlorinated polyethylene is 20~50 wt%.
- 5
7. The incombustible polypropylene resin composition according to claim 1, wherein mean particle size of the antimony trioxide is in the range of 0.2~0.5 μ m.

INTERNATIONAL SEARCH REPORT

International application No.

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A. CLASSIFICATION OF SUBJECT MATTER**IPC7 C08L 23/12, C08K 3/00, C08K 7/14**

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Korean Patents and Applications for Inventions since 1975

Korean Utility Models and Applications for Utility Models since 1975

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

NPS, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 11-21392 A (TAKIRON CO LTD) 26 January 1999 see the whole document	1-7
Y	JP 57-5734 A (TOKUYAMA SODA CO LTD) 12 January 1982 see the whole document	1-7
Y	KR 93-7003488 A (PQ CORPORATION) 30 November 1993 see the whole document	1-7
A	JP 61-176647 A (TOKUYAMA SODA CO LTD) 08 August 1986 see the whole document	1-7
A	JP 08-59902 A (DAI ICHI KOGYO SEIYAKU CO LTD) 05 March 1996 see the whole document	1-7

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Further documents are listed in the continuation of Box C.

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See patent family annex.

* Special categories of cited documents:

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/KR01/01064

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 11-21391 A	26.01.99	none	
JP 57-5734 A	12.01.82	none	
KR 93-7003488 A	30.11.93	US 5409980 A AU 3125593 A1 CA 9309169 A2 EP 565711 A1 GB 565711 R1 JP 6506978 T2 WO 9309169 A2	25.04.95 07.06.93 13.05.93 20.10.93 20.10.93 04.08.94 13.05.93
JP 61-176647 A	08.08.86	none	
JP 08-59902 A	05.03.96	none	